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Combined hydrothermal carbonization and gasification of biomass with carbon capture

B. Erlach*, B. Harder, G. Tsatsaronis

Technische Universität Berlin, Institute for Energy Engineering, Marchstr 18, 10587 Berlin, Germany

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ABSTRACT

Bio-energy with carbon capture and storage (BECCS) can result in negative net carbon emissions and may therefore provide an important technology option for meeting current greenhouse gas stabilization targets. To this end, syngas from biomass gasification combined with pre-combustion carbon capture can be used to produce either biofuels or electricity. Pre-treating the biomass with hydrothermal carbonization (HTC) produces a coal-like substance, biocoal, which is potentially better suited for entrained flow gasification than raw biomass. This paper compares HTC followed by entrained flow gasification of the biocoal with fluidized bed gasification of raw wood, both with carbon capture and storage (CCS). Simulation studies undertaken with Aspen Plus are interpreted using exergy analysis. Syngas production is more efficient from biocoal than from raw wood but the conversion losses in the HTC process outweigh the efficiency gains in the gasification. Carbon losses through gaseous and dissolved byproducts in the HTC also limit the capture rate. A CCS-IGCC with fluidized bed gasification using raw wood results in an electrical efficiency of 28.6% (HHV) and a carbon capture rate of 84.5%, while the conversion chain of HTC and a CCS-IGCC with entrained flow gasification yields an electrical efficiency of 27.7% and a capture rate of 72.7%.

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1. Introduction

Syngas from biomass gasification is widely discussed as an intermediate feedstock for the production of electricity and second-generation biofuels – and increasingly now for bio-energy with carbon capture and storage (BECCS) [1,2]. BECCS may lead to negative carbon emissions if the biomass is grown sustainably. Several studies have found that such negative emissions technologies might be essential for achieving stabilization targets of below 450 ppm CO_2 [3,4].

For coal gasification, with and without CCS, the preferred technology is pressurized oxygen-blown entrained flow gasification. The advantages of entrained flow gasification are a high syngas quality with high H_2 and CO concentrations and low tar and hydrocarbon content, as well as a short residence time resulting in compact gasifier equipment. Several large-scale coal-fed entrained flow gasifiers with a capacity of several hundred MW_{th} are in operation, but none with carbon capture. For biomass, however, pressurized entrained flow gasification is infeasible because of problems related to the feeding system. The gasifier requires feedstock at small particle size (0.1 mm for coal). Laboratory-scale tests with pulverized wood indicated that a pneumatic feeding system is not suitable for wood powder, due to the high cohesion between the fibrous particles [5]. Fluidized bed gasifiers accept a particle size of up to 70 mm [6] and are therefore better suited for biomass. Typical plant input capacities range up to 50 MW_{th} [6]. However, the syngas contains tar and CH_4 and is therefore less suited for carbon capture.

The foregoing observations indicate three options for biomass gasification with CCS:

1. adopt fluidized bed gasification and condition the syngas so that is meets the requirements of the shift reactor and CO_2 removal unit,
2. develop a dedicated biomass feeding system for entrained flow gasifiers,
3. pre-treat the biomass to make it suitable for fine milling and use in the conventional pneumatic transport machinery employed in entrained flow gasifiers.

Research on a dedicated biomass feeding system using screw feeders and piston compressors is presented in Ref. [5]. This system requires particles no smaller than 1 mm, but since biomass

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* Corresponding author. Tel.: +49 30 314 28 449; fax: +49 30 314 21 683.
E-mail address: erlach@iet.tu-berlin.de (B. Erlach).

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is more reactive than coal, bigger particle sizes may be acceptable for gasification.

Several biomass pre-treatment technologies, which convert biomass into a coal-like substance by chemical processing, have been suggested. Examples of such technologies, all in the research and development stage, include torrefaction (a form of mild pyrolysis) [7], the production of a pumpable oil/char slurry by fast pyrolysis [8], and hydrothermal carbonization (HTC). These technologies aim to raise the heating value of the biomass (by increasing the C/O ratio) and to improve its mechanical properties such as grindability [7]. Since they all increase the energy density of the biomass feedstock, they might also benefit the logistics of biomass use. Biomass can be upgraded in decentralized pre-treatment plants and transported to a central power station, thereby avoiding long-distance transport of low density, high moisture biomass. This should also suit the BECCS concept, since carbon capture is more likely to be employed at large facilities.

Some degree of energy densification can also be achieved by drying and pelletizing the biomass, without any chemical conversion. The production of wood pellets is fully commercial, and wood pellets are successfully co-fired in coal-fired power stations [9]. Regarding long-distance transport and co-combustion with coal, wood pellets may be similarly well suited as torrefied wood or HTC biocoal. However, for entrained flow gasification, some form of chemical conversion like torrefaction or HTC is necessary to destroy the fibrous nature of the biomass particles and thereby avoid the problems with the pneumatic feeding systems mentioned above.

In this paper, HTC is investigated as the pre-treatment for biomass gasification with CCS. The HTC reaction takes place in pressurized water at 200–250 °C at or above saturation pressure, with a residence time of 3–8 h. The product, biocoal, is comparable in properties to lignite. Unlike torrefaction and pyrolysis, HTC does not require prior drying and is therefore well suited for processing high moisture biomass. HTC followed by entrained flow gasification with CCS is compared to the direct gasification of wood in entrained flow and fluidized bed gasifiers, also with CCS. These schemes are modelled with the simulation package *Aspen Plus V7.1*. An exergy analysis is performed to identify thermodynamic inefficencies and potentials for improvement.

Section 2 describes the HTC process. Section 3 covers the syngas production from biocoal and wood, via entrained flow and fluidized bed gasification. Section 4 compares a CCS-IGCC with entrained flow gasification using biocoal with a CCS-IGCC with fluidized bed gasification using wood.

2. Biomass pre-treatment with HTC

Hydrothermal carbonization, sometimes referred to as wet torrefaction, is an artificial coalification process which takes place in hot pressurized water between 175 °C and 250 °C. It involves hydrolysis, decarboxylation, dehydratisation, condensation and aromatization reactions [10]. The process was first suggested by Bergius [11] in 1913. Up to the 1960s, the technology was employed in several pilot-scale plants for upgrading peat [12]. The process has, in the last few years, attracted renewed interest for producing improved solid biofuels [13] as well as biochars for soil amendment [14]. A variety of biomass feedstocks, including wood, straw, cut grass, municipal waste, digestate from anaerobic digestion, distiller's grains, microalgae and bark mulch have been successfully carbonized with HTC in laboratory-scale experiments [15–19]. The degree of carbonization depends on the reaction temperature and residence time, where higher temperatures and longer residence times lead to a higher carbon content and calorific value, but also to lower mass and energy yields [20]. Aside from biocoal, HTC produces water, CO_2, small amounts of CO, H_2 and hydrocarbons, and dissolved organic and inorganic compounds. Gerhardt, Berg and Kamm identified acetic acid, formic acid, glycolic acid, levulinic acid, phenol, furfural, HMF, and sugars in the aqueous phase using HPLC analysis [21]. They also found that 29% of the total organic carbon in the aqueous phase has not been identified, indicating that more carbon containing species are present. Organic acids account for approximately 75% of the identified products.

By destroying the structure of the biomass and making it more hydrophobic, HTC facilitates mechanical dewatering. Dry matter contents of 57–68% were achieved with laboratory presses for biocoal from organic waste [22]. Efficient mechanical dewatering reduces the energy requirement for thermal drying of the biocoal.

2.1. HTC plant model

A flow sheet and *Aspen Plus* simulation results for an industrial-scale HTC plant with a continuous reactor have been presented earlier [20,23]. The process design was based on a plant for hydrothermal upgrading of peat which was operated in Sweden around 1960 [24]. In this paper, the flow sheet and simulations are further developed and an exergy analysis is presented. The process flow diagram is shown in Fig. 1. Biomass is mixed with recycled process water to create a pumpable slurry (stream 2), and pressurized in 4 steps with the slurry pumps (P1–P4). The slurry is

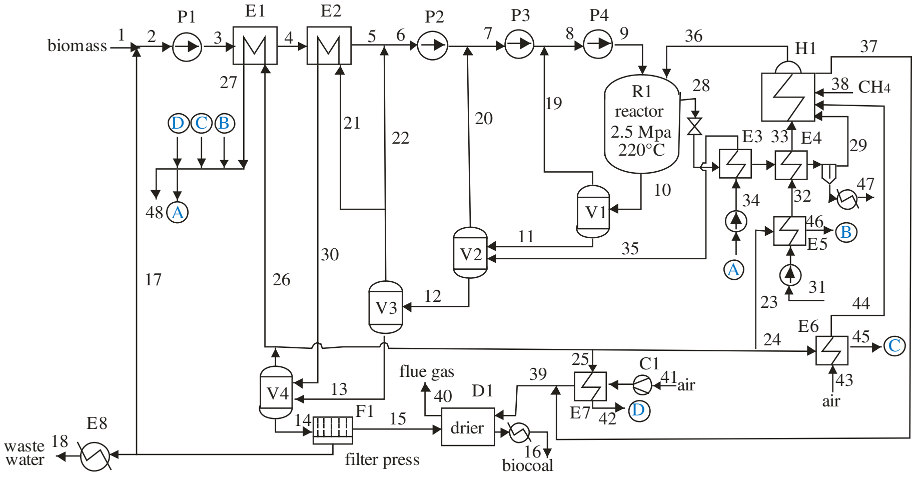

Fig. 1. Flow sheet of the HTC plant.

preheated in several steps, first by heat exchangers (E1, E2), then by mixing with steam recovered at four pressure levels by flashing the reaction products in the flash tanks (V1–V4). The additional steam required to reach the reaction temperature is produced by a natural gas fired boiler (H1). The combustion air is preheated with steam (E6). The gaseous reaction products, which contain CO and other combustible compounds, are also fed to the boiler. Since they are saturated with water vapour, they are first cooled to condense part of this water. Recovered heat from the gaseous byproducts is used for steam production (E3) and feedwater preheating (E4, E5). The biocoal, after de-pressurizing, is mechanically dewatered by a filter press (F1) and dried (D1). Low temperature drying is employed, with heat provided by the exhaust gas from the boiler and steam recovered at 0.1 MPa (E7). The waste water is cooled (E8) and submitted to further treatment (not considered in the simulation).

2.1.1. *HTC reaction*

The HTC reactor is modelled as a black box, with biomass and biocoal composition based on laboratory experiments with poplar wood, using a temperature of 220 °C and a residence time of 4 h [25]. The composition of the gaseous byproducts of the HTC reactor, comprising 91.8% CO_2, 7.1% CO, 1.0% H_2 and 0.1% CH_4, is also based on these experiments. The bulk of these experiments were conducted with fresh water, rather than recycled process water, and using a high water to biomass ratio of 9:1, neither of which would be representative for an industrial plant. Dissolved organics amounted to 16–17% of the feedstock biomass (by mass) in these experiments. Experiments conducted in process water recovered from previous experiments indicated lower dissolved organics losses (5–9%). For the simulation, we assume that 10% of the biomass feedstock is converted to dissolved organics. Biomass, biocoal and dissolved compounds were modelled based on their elemental composition. Higher heating values (HHV) were calculated using a correlation from Channiwala [26]. The elemental composition of the dissolved organics is based on the results of HPLC analysis [25]. H_2O and gaseous reaction products were then calculated by difference from the elemental balances in the simulation, resulting in a gas yield of 6.3% of the dry biomass. For comparison, measured gas yields ranged between 4 and 10%. Based on the experimental results, 75% of the inorganic matter of the biomass is taken to be dissolved in the process water.

In the drier, the dissolved inorganic and part of the dissolved organic compounds recondense on the biocoal. Since the organic compounds are modelled using their elemental composition only, simulation results do not indicate how much of these fractions evaporate in the flash stages and the drier. We assumed that 40% recondenses on the coal and that the rest is lost via the drier exhaust and condensate from the flash steam. The recondensation in the drier leads to a slight change in the composition of the biocoal. Elemental compositions, higher heating values and mass and energy yields γ_m and γ_e, defined by (1), where $\dot{m}$ is the mass flow rate, are shown in Table 1.

$$\gamma_e = \gamma_m \frac{\mathrm{HHV}_{\mathrm{biocoal,dry}}}{\mathrm{HHV}_{\mathrm{biomass,dry}}} = \frac{\dot{m}_{\mathrm{biocoal,dry}}\mathrm{HHV}_{\mathrm{biocoal,dry}}}{\dot{m}_{\mathrm{biomass,dry}}\mathrm{HHV}_{\mathrm{biomass,dry}}} \quad (1)$$

2.1.2. *Operating parameters and energy balance*

This section describes the selected and computed operating parameters and the energy balance for an HTC plant with 11.15 $\mathrm{MW_{HHV}}$ input capacity using poplar wood. Table 2 shows the temperatures, pressures and mass flow rates of the key streams. The following assumptions regarding heat losses and efficiencies are used in the simulation:

- The slurry pump efficiency is 45%.
- The heat loss of the reactor is 5 kW, equivalent to 0.05% of the biomass energy. Heat losses of biomass slurry and biocoal slurry are 13 kW and 12 kW, respectively, corresponding to 1 °C of cooling.
- Boiler and drier heat losses are 3% and 5%, respectively.
- The drier exhaust temperature is 10 °C above condensation temperature.
- Minimum heat exchanger temperature difference is 10 °C.
- The pressure difference between flash steam and slurry in the mixing preheaters is 2 bar.
- The dry matter content (by mass) is 15% in the biomass slurry (stream 2) and 60% after mechanical dewatering.
- Natural gas is modelled as CH_4.

Table 1
Characteristics of wood, biocoal and dissolved organic compounds.

	Poplar wood	Biocoal, reactor outlet	Biocoal, drier outlet	Dissolved organics, reactor outlet
Carbon	49.75%	64.88%	63.83%	40.52%
Hydrogen	6.08%	5.70%	5.66%	5.36%
Oxygen	42.85%	28.93%	29.40%	54.12%
Ash	1.32%	0.49%	0.00%	0.00%
Water (wet basis)	50.00%		10.35%	0.00%
HHV [MJ/kg]	20.070	26.366	25.884	14.864
Specific chemical exergy (dry basis) [MJ/kg]	21.094	27.221	26.736	15.855
Mass yield (dry basis)		67.7%	70.0%	10.0%
Energy yield (HHV)		88.9%	90.2%	7.4%

The scheme produces 10.06 $\mathrm{MW_{HHV}}$ of biocoal from a biomass input of 11.15 $\mathrm{MW_{HHV}}$. The auxiliary energy consumption per MJ of biocoal amounts to 965 kW of natural gas for the boiler and 95 kW of electricity for slurry pumps (37 kW), drier air fan (37 kW), filter press (19 kW) and pumps (2 kW).

The overall energetic efficiency η of the HTC plant, defined by (2), is 82.42%.

$$\eta = \frac{\dot{m}_{\mathrm{biocoal}}\mathrm{HHV}_{\mathrm{biocoal}}}{\dot{m}_{\mathrm{biomass}}\mathrm{HHV}_{\mathrm{biomass}} + \dot{m}_{\mathrm{CH_4}}\mathrm{HHV}_{\mathrm{CH_4}} + \dot{W}} \quad (2)$$

Table 2
Mass flow (*m*), temperature (*T*) and pressure (*p*) of key flow streams for the HTC plant.

No.	Material	*m* [kg/s]	*T* [°C]	*p* [bar]
1	Biomass	1.1111	15	1.01
5	Biomass slurry	3.9910	100	3.00
9	Biomass slurry	4.7357	188	27.00
14	Biocoal slurry	3.8685	100	1.01
16	Biocoal	0.4336	15	1.01
18	Waste water	0.3340	40	1.01
19	Steam	0.1907	204	17.00
20	Steam	0.3186	184	11.00
21	Steam	0.0736	152	5.00
22	Steam	0.2355	152	5.00
23	Steam	0.0476	100	1.01
24	Steam	0.0102	100	1.01
25	Steam	0.2308	100	1.01
26	Steam	0.1168	100	1.01
28	Reactor offgas[a]	0.2211	220	23.00
36	Steam	0.3929	232	29.00
37	Boiler exhaust	0.4067	208	1.01
38	Natural gas	0.0170	15	1.01
40	Drier exhaust	8.6681	45	1.01
48	Condensate	0.2942	62	1.01

[a] 92.67% H_2O, 6.72% CO_2, 0.52% CO, 0.07% H_2, 0.01% CH_4.

2.2. Exergy analysis

An exergy analysis was conducted using the chemical exergy model of Szargut [27]. The chemical exergy of ash and dissolved inorganic compounds is neglected. Table 3 shows the exergy of the fuel and product streams, the exergy losses $\dot{E}_L$ related to waste streams, and the exergy destruction within the plant components$\dot{E}_D$. The exergetic efficiency ε of the HTC plant, defined by (3), is 81.4%.

$$\varepsilon = \frac{\dot{E}_{\text{biocoal}}}{\dot{E}_{\text{biomass}} + \dot{E}_{\text{CH}_4} + \dot{E}_{\text{air}} + \dot{E}_{\text{H}_2\text{O}} + \dot{W}_{\text{el}}} \tag{3}$$

The main sources of exergy destruction and loss are the HTC reactor itself, the steam generator, the drier and the organic compounds exported with the waste water, condensate and drier effluent. The losses of organic compounds amount to 31% of the total $E_D + E_L$ or 6% of the exergetic fuel of the plant. It should be noted that there is a significant uncertainty about this value, since the behaviour of a continuous flow reactor with recycled process water and flash cooling might be rather different from the bench-scale experiments with fresh water and slow cooling of the reaction products. Further research on the reaction chemistry is required to find out whether the formation of dissolved compounds can be limited by choosing the operating conditions accordingly. Options to convert the dissolved organics into useful products should also be investigated. Experiments with anaerobic degradation indicate that biogas production from HTC waste water could be promising [22]. Another approach is the separation and recovery of high value chemicals, such as phenol and acetic acid. In this case, reaction conditions might need to be modified to enhance the formation of the desired compounds. The exergy destruction from the HTC reaction itself amounts to 17% of total $E_D + E_L$. That said, the influence of the operating parameters on the reaction chemistry warrants further exploration and may offer improvements in yield.

The boiler, with an exergetic efficiency of only 39.5%, accounts for 22% of total $E_D + E_L$, more than the HTC reactor. Exergy destruction in the boiler can be reduced by limiting the steam requirement. Decreasing the water to biomass ratio in the slurry decreases the amount of additional steam needed [20], but the extent to which this is possible depends on the technical requirements of the feeding system. The drier accounts for 9% of total $E_D + E_L$, but is fuelled by low temperature waste heat, namely steam at 100 °C and the boiler exhaust gas, which otherwise presents few opportunities for reuse.

Table 3
Exergy of fuel, product and waste streams and exergy destruction of the HTC plant.

Exergetic fuel	***E* [kW]**
Biomass	11,747
Natural gas	882
Electricity	95
Air	38
Water	20
Total	**12,782**
Exergetic product	
Biocoal	**10,402**
Exergy losses (E_L)	
Exhaust gas	96
Waste water (chemical exergy)	496
Other organics losses	240
Waste water (physical exergy)	2
Condensate	29
Total	**863**
Exergy destruction (E_D)	
HTC reactor	394
Drier	209
Boiler	530
Slurry pumps	28
Slurry preheaters (indirect)	22
Slurry preheaters (mixing)	50
Reactor gas heat exchangers	29
Flash tanks	153
Coolers	57
Piping heat losses	19
Filter press	19
Other	8
Total	**1518**

3. Syngas production with CCS

This section describes the syngas production from wood chips with CCS via three pathways:

- pre-treatment of the wood chips with HTC followed by entrained flow gasification (*HTC-EF-biocoal*)
- direct entrained flow gasification of the wood chips (*EF-wood*)
- fluidized bed gasification of the wood chips (*FB-wood-1*, *FB-wood-2*, *FB-wood-3*)

For case *HTC-EF-biocoal*, it is assumed that biocoal is produced in several decentral HTC plants and gasified in a central plant with a biocoal input capacity of 2250 MW_{HHV}. Transport emissions are neglected in the calculations. Fluidized bed gasification is assumed to have a wood chips input capacity of 480 MW_{HHV}.

3.1. Entrained flow gasification

Entrained flow gasification of coal with CCS has been widely analyzed using simulation studies, for example [28–30]. Since biocoal has similar properties to fossil coal, a typical plant arrangement for coal gasification with CCS is employed. A flow sheet of the process is shown in Fig. 2. The biocoal is milled, dried to 5% water content and pressurized with lock hoppers. The oxygen for the gasifier is produced in a cryogenic air separation unit (ASU). The raw gas leaving the gasifier is cooled to below the slag solidifying temperature (900 °C) by mixing with a colder raw gas recycle stream. It is then further cooled and cleaned in a wet scrubber. A two stage sour shift reactor converts CO to CO_2 and, after cooling to 40 °C, sulphur components and CO_2 are removed by physical absorption with the solvent Selexol in the acid gas removal unit (AGR). The separated CO_2 is compressed to 110 bar for transport and sequestration. The waste heat from gas cooling at several stages is used to produce steam at various pressure levels and for feedwater preheating. Properties of the key flow streams are given in Table 4.

The most important modelling assumptions are as follows:

- The gasifier carbon conversion efficiency is 99.7%.
- The syngas composition is calculated at chemical equilibrium.
- The CO conversion rate of the sour shift reactor is 94%.
- The AGR is modelled as a black box with a CO_2 removal rate of 90%, a heat demand of 4590 kJ/kmol and an electricity demand of 3149 kJ/kmol raw gas, based on literature data [29]. CO_2 is recovered from the AGR at 23 bar, 9 bar and 3 bar.
- Gasifier and shift reactor heat loss are each 2% of fuel energy.
- Heat losses in percent of fuel energy: gasifier 2%, shift reactor 2%, drier 5%.

We also modelled the entrained gasification of wood (*EF-wood*), implying that the required feeding technology, comprising screw

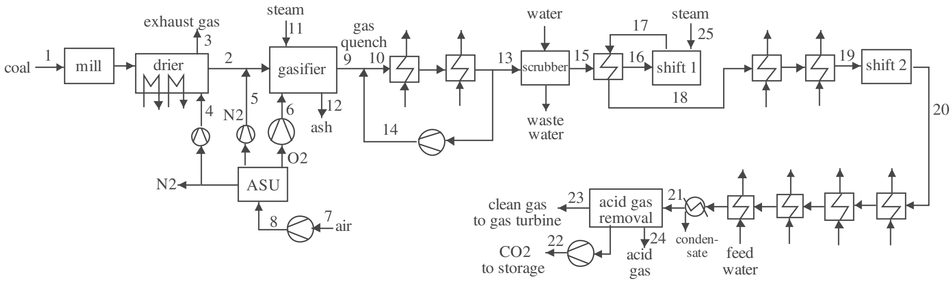

Fig. 2. Flow sheet of the entrained flow gasification process with carbon capture (*EF-biocoal*).

feeder and piston compressor as described in [5] has been developed. The flow sheet is identical to Fig. 2, except that the wood is first dried to 10% water content, and then milled, because the energy requirement for milling wood is strongly dependant on its moisture content. Low pressure saturated steam at 105 °C is used to fuel the drier.

3.2. *Fluidized bed gasification*

The operating temperature of fluidized bed gasification is limited to 900 °C or lower by the ash melting temperature of the biomass. Such temperatures lead to tar and hydrocarbon formation. Tar is a much discussed problem for biomass gasification and different strategies for tar removal are described in the literature, for example [31]. In a process with CCS, the shift catalysts have to be protected from poisoning, which increases the requirements for gas cleaning [1].

Table 4
Mass flow (*m*), temperature (*T*) and pressure (*p*) of key flow streams in the entrained flow gasification process with carbon capture (*EF-biocoal*).

No	Type	*m* [kg/s]	*T* [°C]	*p* [bar]
1	Biocoal	97.29	15	1.01
3	Drier exhaust[a]	85.53	60	1.01
6	Oxygen[b]	66.24	30	43.00
8	Air	297.38	30	4.60
9	Raw gas	174.69	1550	39.00
10	Raw gas	390.10	900	39.00
11	Steam	5.51	253	43.00
13	Raw gas	174.69	310	37.51
15	Raw gas[c]	192.76	167	36.01
16	Raw gas	192.76	275	35.73
19	Raw gas	303.50	247	33.30
20	Raw gas	303.50	341	31.30
21	Raw gas[d]	225.85	38	30.48
22	CO_2[e]	173.29	40	110.00
23	Clean gas[f]	52.53	34	27.20
25	Steam	110.74	253	43.00

[a] 88.3% N_2, 1.9% O_2, 9.8% H_2O.
[b] 95% O_2, 5% N_2.
[c] 44.6% CO, 21.8% H_2, 22% H_2O, 5.8% CO_2, 5.9% N_2.
[d] 53.0% H_2, 39.6% CO_2, 2.2% CO, 4.9% N_2, 0.2% H_2O.
[e] 96.9% CO_2, 2.6% H_2, 0.2% CO, 0.2% H_2O, 0.1% N_2.
[f] 82.4% H_2, 6.3% CO_2, 3.4% CO, 7.7% N_2, 0.2% H_2O (mol%).

Formation of methane and other light hydrocarbons is not regarded an issue for IGCC without CCS, since these have a high calorific value and can be burned in the gas turbine system. If pre-combustion carbon capture is applied, however, hydrocarbons pass through the shift reactor without being converted, and thus escape downstream carbon capture [1]. Because of the low H_2S concentration in the raw gas from wood, solid sorption may prove suitable for sulphur removal [32].

Fig. 3 shows the flow sheet of a fluidized bed gasification process with pre-combustion carbon capture. The wood chips are dried to 10% water content and fed to the pressurized gasifier, where they are gasified in a sand bed with a steam/oxygen mixture. Dolomite is used to keep tar formation below 2 g/m^3 (STP), in order to prevent coke formation in the steam reformer [33]. Heat for the wood drier is supplied by low pressure steam. Tar and methane are converted to CO and H_2 in a catalytic steam reforming process using a nickel-based catalyst [34]. Since the catalysts have been found to be more resistant against sulphur at high temperatures [35], the steam reformer is operated at 950 °C. Thermal energy for the endothermic steam reforming reaction is provided by burning part of the clean gas. The combustion gas leaves the steam reformer at 970 °C and can be used to raise steam in the heat recovery steam generator. The desulfurization with ZnO sorbents [36] takes place in two beds at 480 °C and 400 °C (bed-1, bed-2). Since the elemental composition of the wood is modelled without sulphur, the desulfurization process has no effect on the simulation results. A two stage clean shift followed by CO_2 removal with the solvent Rectisol are applied. Further details regarding the flow sheet design are given in [37].

No complete set of measured data for the pressurized gasification of wood in an oxygen/steam mixture was found in literature. Therefore, a gasification model was calibrated based on experimental data from [33] for an oxygen/steam blown gasifier under atmospheric conditions. Temperature difference parameters for the

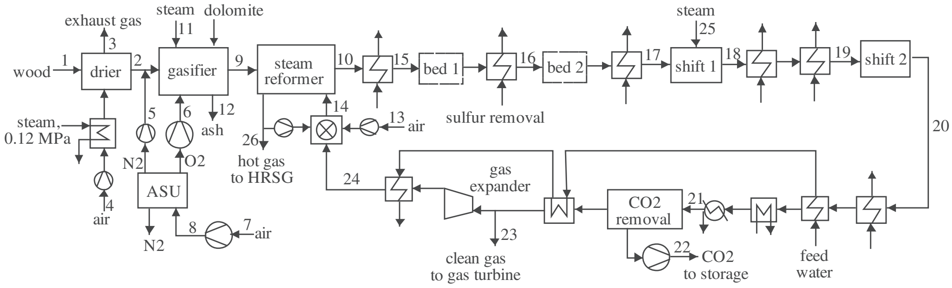

Fig. 3. Flow sheet of the fluidized bed gasification process with carbon capture (*FB-wood-1*).

gasification reactions were adjusted to match the syngas composition from [33]. The same temperature difference parameters were then used to model the pressurized gasification, where the steam per biomass ratio was held constant and the oxygen was adjusted to obtain the desired gasification temperature of 835 °C. This approach seems to result in unrealistically high methane concentrations. Therefore, a second simulation case was run (*FB-wood-2*), where equilibrium conditions were applied to all gasification reactions. This yields an unrealistically low methane concentration, since chemical equilibrium is usually not reached in fluidized bed gasification. The syngas methane concentrations in the simulations *FB-wood-1* and *FB-wood-2* can therefore be considered upper and lower bounds, the real syngas composition is likely to be somewhere in between.

The most important modelling assumptions are as follows:

- Biomass water content is 50% as delivered and 10% after drying.
- The gasifier carbon conversion efficiency is 98%.
- Tar is modelled as phenol, toluene and pyrene, the raw gas tar content being 1.65 g/m^3 (STP).
- The steam reforming reaction is calculated using chemical equilibrium.
- The CO conversion rate of the clean shift reactor is 98.5%.
- The CO_2 absorption is modelled as a black box with a capture rate of 90%, a heat demand of 1045 kJ/kmol and an electricity demand of 1678 MJ/kmol raw gas, based on [28]. CO_2 is recovered at three pressure levels: 10.0 bar, 2.7 bar and 1.4 bar.
- Heat losses as a percent of fuel energy: gasifier 3%, reformer 3%, shift reactor 2%, drier 5%.

The properties of key flow streams for case *FB-wood-1* are given in Table 5. The temperatures and pressures in cases *FB-wood-2* are largely the same as in *FB-wood-1*, while the mass flow rates and compositions vary.

We assume in the simulation cases *FB-wood-1* and *FB-wood-2* that the wood chips are dried with low temperature steam. However, high temperature drying with gas temperatures of 600–1000 °C is more widely employed for large-scale biomass gasification [38]. Therefore, an additional simulation case *FB-wood-3* was considered, whereby the hot gas stream leaving the steam reformer is used to dry the wood chips. Operating data for case *FB-wood-3* is identical to *FB-wood-1* for all process units except the drier. The results for all three cases are discussed in the following sections.

Table 5
Mass flow (m), temperature (T) and pressure (p) of key flow streams in the fluidized bed gasification process with carbon capture (*FB-wood-1*).

No	Type	m [kg/s]	T [°C]	p [bar]
1	Wood	47.85	15	1.01
3	Drier exhaust[a]	1259.09	41	1.01
6	Oxygen	4.96	30	37.30
8	Air	22.12	30	4.60
9	Raw gas[b]	44.76	835	31.30
10	Raw gas[c]	44.76	950	29.30
11	Steam	13.64	360	37.30
14	Exhaust gas	100.50	1601	1.05
15	Raw gas	44.76	480	28.40
17	Raw gas	44.76	400	26.26
19	Raw gas	61.79	200	23.39
20	Raw gas	61.79	217	21.04
21	Raw gas[d]	45.28	30	20.72
22	CO_2[e]	36.85	31	110.00
23	Clean gas[f]	4.87	145	17.02
24	Clean gas	3.48	20	1.50
25	Steam	17.03	350	26.67
26	Exhaust gas	44.76	480	28.40

[a] 76.1% N_2, 20.2% O_2, 3.6% H_2O.
[b] 20.2% CH_4, 6.2% CO, 5.5% H_2, 41.7% H_2O, 25.7% CO_2, 0.7% N_2.
[c] 1.3% CH_4, 25.6% CO 37.1% H_2, 24.2% H_2O, 11.3% CO_2, 0.5% N_2.
[d] 1.3% CH_4, 0.4% CO, 61.6% H_2, 0.2% H_2O, 36.1% CO_2, 0.5% N_2.
[e] 97.2% CO_2, 2.6% H_2, 0.2% CO.
[f] 91.4% H_2, 5.4% CO_2, 0.5% CO, 0.7% N_2, 1.9% CH_4 [mol%].

3.3. Simulation results

Key results for the gasification of wood and biocoal in the various simulation cases are given in Table 6. Final clean gas yields and the auxiliary energy consumption are given in Table 7, normalized to 100 MW_{HHV} of gasifier feedstock input. Simulation results for the entrained flow gasification of bituminous coal using the same process configuration are included for comparison.

The CH_4 concentration in the raw gas of the fluidized bed gasification is strongly dependent of the simulation model used for the gasification. *FB-wood-1* yields a surprisingly high methane concentration of 20.3%, while case *FB-wood-2* based on chemical equilibrium leads to a CH_4 concentration of only 3.5%. This corresponds to 28.4% and 6.5% of the feedstock carbon for *FB-wood-1* and *FB-wood-2* respectively, which would bypass the capture without downstream steam reforming. The oxygen demand of the gasifier is almost double in *FB-wood-2*, as the endothermic steam reforming reaction largely takes place in the gasifier itself rather than in the downstream catalytic reactor.

Entrained flow gasification has a higher oxygen demand due to the higher gasification temperature. This results in a lower cold gas efficiency $(\dot{m}_{\text{gas}}\text{HHV}_{\text{gas}}(\dot{m}_{\text{feed}}\text{HHV}_{\text{feed}})^{-1})$ when compared to fluidized bed gasification. The exergetic efficiency of the gasifier, however, lies in the same range for both gasification processes. This is due to the higher temperature and associated physical exergy of the entrained flow gasifier raw gas, which is accounted for as part of the product by the exergetic efficiency, but is neglected by the definition for the cold gas efficiency.

For the entrained flow gasification, the feedstock has a significant influence on the exergetic efficiency, which results in 80.3% for wood, 82.8% for biocoal and 85.8% for bituminous coal. The lower the calorific value of the feedstock, the higher is the proportion of fuel that needs to be oxidized to reach the gasification temperature, resulting in a higher oxygen demand and higher CO_2 and H_2O concentrations in the syngas. Despite the higher cold gas efficiency of fluidized bed gasification, the net yield of clean gas is 16% lower than for the entrained flow gasification of wood (*EF-wood-1*) in the simulation cases with high CH_4 production, because 42% of the clean gas is burned to provide heat for the steam reformer.

The electricity consumption is higher in the entrained flow gasification cases, due to the higher oxygen demand requiring compression work and because of feedstock milling and pressurizing. All cases except *FB-wood-2* have a net export of heat in the form of steam and hot gas from the reformer as a byproduct, which can be used in a heat recovery steam generator. In the cases with wood as the gasification feedstock, the biggest consumer of thermal energy is the drier, with 14–19 MW per 100 MW of biomass feedstock.

3.4. Exergy analysis

Results from the exergy analysis are first discussed for the syngas production process, and then for the conversion chain of HTC followed by gasification. For the conversion pathway via biocoal, *HTC-EF-biocoal* denotes the overall conversion chain, while *EF-biocoal* refers to the syngas production only. Table 8 shows the exergy of the input and output streams, the exergy losses $\dot{E}_L$ related to waste streams, and the exergy destruction within the plant components $\dot{E}_D$.

Table 6
Gasification agents, composition and higher heating value of the raw gas, and efficiency of the gasifier.

		FB-wood-1	*FB-wood-2*	*FB-wood-3*	*EF-wood*	*EF-biocoal*	EF-coal
Oxygen	[kg/kg feed]	0.186	0.364	0.185	0.550	0.722	0.840
Steam	[kg/kg feed]	0.513	0.513	0.513	0.060	0.060	0.060
CO_2	[mol%]	25.74%	18.94%	25.72%	11.98%	6.49%	1.87%
CO	[mol%]	6.16%	17.15%	6.16%	38.11%	50.06%	59.52%
H_2	[mol%]	5.50%	27.54%	5.50%	21.84%	24.50%	27.84%
H_2O	[mol%]	41.69%	32.01%	41.64%	26.71%	12.36%	3.40%
CH_4	[mol%]	20.21%	3.51%	20.27%	0.00%	0.00%	0.01%
N_2	[mol%]	0.68%	0.82%	0.67%	1.37%	6.58%	7.05%
H_2S	[mol%]						0.31%
Tar	[mol%]	0.03%	0.03%	0.03%	0.00%	0.00%	0.00%
Cold gas efficiency	[−]	82.8%	81.1%	82.9%	69.9%	76.7%	80.6%
Exergetic efficiency	[−]	84.3%	80.6%	84.4%	80.3%	82.8%	85.8%
Raw gas HHV (ar)	[MJ/kg]	8.883	7.870	8.908	7.890	9.874	12.041

We define the exergetic efficiency ε of syngas production as:

$$\varepsilon = \frac{\dot{E}_{\text{clean gas}} + \dot{E}_{\Delta\dot{Q},\text{net}}}{\dot{E}_{\text{biomass}} + \dot{E}_{\text{air}} + \dot{E}_{\text{steam}} + \dot{E}_{\text{water}} + \dot{W}_{\text{el}} + \dot{E}_{\text{sand}} + \dot{E}_{\text{dolomite}}} \quad (4)$$

where subscript $\Delta\dot{Q}_{\text{net}}$ relates to the thermal energy in the form of steam and hot water which is generated in the raw gas coolers, plus the thermal energy of the reformer exhaust gas (stream 26 in Fig. 3) minus the thermal energy consumed by air separation unit, acid gas removal and drier. $\dot{E}_{\text{steam}}$ and$\dot{E}_{\text{water}}$ refer to the steam used as gasification agent and the water for the scrubber. $\dot{E}_{\text{sand}}$ is the make-up for the fluidized bed material lost with the ash, and $\dot{W}_{\text{el}}$ is the net electricity input. The captured and compressed CO_2 stream is *not* considered an exergetic product of the plant, although for biomass with CCS the negative net emissions constitute the raison d'être of the plant.

The total exergetic efficiency of the syngas production process is 52.4–59.0% for wood, 65.1% for biocoal and 67.7% for bituminous coal. The efficiency of the fluidized bed gasification is strongly dependent on the methane concentration in the raw gas. In case *FB-wood-2* with low methane, the exergetic efficiency is higher than for the entrained flow gasification of wood, *EF-wood-1*. For case *FB-wood-1* with a high methane concentration, the exergetic efficiency is 1.8 percentage points lower than for *EF-wood-1* because of the large portion of syngas being burned in the reformer. The reformer operates with a relatively low exergetic efficiency of 56%. The gasifier and reformer are the largest sources of exergy destruction, accounting for 38–42% of the total exergy destruction and loss $E_D + E_L$ in the entrained flow cases and 48–52% in the fluidized bed cases. The exergetic efficiency of *FB-wood-3*, using high temperature drying with the reformer exhaust gas, is 3.8 percentage points lower than that of case *FB-wood-1* where low temperature drying is employed. The drier and drier exhaust gas together account for 13–19% of $E_D + E_L$ in the cases where wood is gasified.

The syngas production from biocoal is more efficient than that from wood. However, the overall conversion chain of HTC and subsequent biocoal gasification, taking into account the losses and auxiliary energy consumption of the HTC process, yields an exergetic efficiency of 54.4%. This is better than *FB-wood-3* with a high methane concentration in the raw gas and high temperature drying, but worse than the other cases with direct gasification of the wood chips using fluidized bed gasification. Viewed separately, 36% of the overall exergy destruction and losses take place in the HTC plant and 64% in the gasification process.

3.5. Carbon performance

For BECCS systems, the carbon capture rate might be equally or even more important than the energetic and exergetic efficiencies. Table 9 shows the carbon balance for the respective simulation cases. While cases with direct gasification of wood result in 84%–86% of the feedstock carbon being captured, *HTC-EF-biocoal-1* drops to 73%. This is mainly due to the natural gas consumption of the HTC plant and the fact that 10% of the biomass carbon is lost in the form of liquid and gaseous byproducts of the HTC process. If the liquid byproducts are oxidized in a treatment plant, their carbon

Table 7
Energy balance for the syngas production, normalized to 100 MW_{HHV} of gasifier feed.

		FB-wood-1	*FB-wood-2*	*FB-wood-3*	*EF-wood*	*HTC-EF-biocoal*	EF-coal
Pre-treatment							
Biomass	[MW]					110.81	
Electricity	[MW]					0.94	
Natural gas	[MW]					9.59	
Syngas production							
Gasifier feed, HHV	[MW]	100.00	100.00	100.00	100.00	100.00	100.00
Net clean gas production	[MW]	58.13	70.15	58.25	68.91	75.71	79.19
Electricity consumption							
Air separation unit	[MW]	1.47	2.87	1.46	4.19	4.05	4.06
AGR and CO_2-compression	[MW]	2.97	3.03	2.97	3.12	3.22	3.18
Milling and pressurizing	[MW]	0.05	0.05	0.05	1.67	2.60	0.65
Air, flue gas and recycle comp.	[MW]	1.58	1.20	0.95	1.19	0.22	0.20
Syngas expander	[MW]	−0.65	−0.22	−0.65	0.00	0.00	0.00
Net total	[MW]	5.41	6.93	4.77	10.17	10.09	8.08
Thermal energy delivered to heat recovery steam generator							
Net steam/hot water production	[MW]	−10.70	−7.78	8.39	3.36	14.02	9.34
Reformer exhaust gas	[MW]	22.00	7.28	0.00	0.00	0.00	0.00
Net heat to HRSG	[MW]	11.30	−0.51	8.39	3.36	14.02	9.34

Table 8
Exergy balance for the syngas production, normalized to 100 MW_{ex} of gasifier feed exergy.

		FB-wood-1	*FB-wood-2*	*FB-wood-3*	*EF-wood*	*HTC-EF-biocoal*	EF-coal
Pre-treatment							
Biomass	[MW_{ex}]					112.93	
Electricity	[MW_{ex}]					0.913	
Natural gas	[MW_{ex}]					8.47	
Air and water	[MW_{ex}]					0.56	
Syngas production							
Fuels	**[MW_{ex}]**	**114.21**	**114.08**	**112.47**	**114.79**	**115.97**	**115.31**
Gasifier feed	[MW_{ex}]	100.00	100.00	100.00	100.00	100.00	100.00
Electricity	[MW_{ex}]	5.18	6.63	4.56	9.77	9.76	7.90
Gasification agents and air	[MW_{ex}]	9.03	7.45	7.91	5.02	6.20	7.41
Products	**[MW_{ex}]**	**64.22**	**67.32**	**58.98**	**66.57**	**75.47**	**78.08**
Syngas	[MW_{ex}]	47.85	57.64	47.95	57.14	63.24	66.73
Steam	[MW_{ex}]	6.85	6.53	11.03	9.43	12.23	11.35
Reformer exhaust gas	[MW_{ex}]	9.52	3.15	0.00	0.00	0.00	0.00
Exergy losses	**[MW_{ex}]**	**10.25**	**10.43**	**12.00**	**8.98**	**7.94**	**8.69**
CO_2	[MW_{ex}]	5.98	6.10	5.97	6.11	6.08	5.91
Slag/ash	[MW_{ex}]	2.03	2.03	2.03	0.40	0.36	1.20
Drier exhaust	[MW_{ex}]	1.98	1.98	3.73	1.94	0.12	0.11
Other	[MW_{ex}]	0.27	0.31	0.26	0.54	1.38	1.48
Exergy destruction	**[MW_{ex}]**	**39.72**	**36.33**	**41.48**	**39.24**	**32.58**	**28.54**
Gasifier	[MW_{ex}]	15.39	18.95	15.28	19.57	17.10	13.99
Steam reformer	[MW_{ex}]	10.76	3.40	10.77	0.00	0.00	0.00
Gas quench and recycle compressor	[MW_{ex}]	0.00	0.00	0.00	1.53	1.26	1.07
Shift reactors	[MW_{ex}]	1.57	1.21	1.57	1.94	2.67	3.12
Air separation unit	[MW_{ex}]	1.04	2.03	1.03	2.92	2.84	2.88
Wood/coal drier	[MW_{ex}]	4.50	4.50	6.26	4.53	1.08	0.50
Steam and hot water generation	[MW_{ex}]	3.53	3.44	3.53	3.88	3.52	3.25
AGR and CO_2-compression	[MW_{ex}]	2.33	2.40	2.33	3.01	3.16	3.16
Other	[MW_{ex}]	0.60	0.38	0.70	1.88	0.94	0.56
Exergetic efficiency, syngas production	**[-]**	**56.2%**	**59.0%**	**52.4%**	**58.0%**	**65.1%**	**67.7%**
Exergetic efficiency, conversion chain	**[-]**					**54.4%**	

content will be released to the atmosphere as CO_2. For the fluidized bed gasification, 2% of the feedstock carbon remains in the ash, while for the entrained flow gasification, 0.3% ends up in the gasifier slag.

4. Biomass CCS-IGCC plants

IGCC plants with CCS were simulated for the cases *FB-wood-1* and case *EF-biocoal*, supplementing the above described syngas production with a gas turbine combined cycle. The combined cycle is modelled with the simulation packages *GateCycle Version 5.52.0r* and *EBSILON Professional 9.00*. Key data is listed in Table 10. For comparison, simulation results for a similar biomass CCS-IGCC configuration by Rhodes [2] are included.

The gas turbine system is modelled using technical data for an F-class heavy-duty turbine [39] for case *EF-biocoal-1* and for a Siemens SGT-600 turbine [40] for *FB-wood*-1. Both types of gas turbine systems have been tested with hydrogen rich gas [41,42]. The gas turbine systems were first simulated fuelled with natural gas in order to adjust the models to the published operating data. The efficiencies for natural gas operation are 35.2% and 31.1% (HHV), respectively. For syngas operation, the syngas is assumed to be diluted with nitrogen and water vapour to a hydrogen content of 53% before combustion for NO_X control. In case *FB-wood*-1, the hot exhaust gas from the steam reformer (stream 26 in Fig. 3) is mixed with the gas turbine exhaust gas. This raises the gas inlet temperature of the heat recovery steam generator (HRSG) to 652 °C, enabling a high live steam temperature of 610 °C.

The energetic efficiency of the biocoal IGCC is 33.3%, 4.7 percentage points higher than for the wood IGCC, due to a bigger and more efficient gas turbine system and for the reasons related to the gasification system discussed in section 3. When fired with bituminous coal, the efficiency of an entrained flow IGCC with the same configuration resulted in 34.9% [43].

Taking into account the prior conversion losses of the HTC process, however, the efficiency of the overall system is 27.7%, lower than the efficiency of the direct gasification of wood. Moreover, the carbon capture rate of HTC with subsequent entrained flow gasification is only 72.7%.

The biomass CCS-IGCC analyzed in [2] has a similar efficiency to our model, but a much lower carbon capture rate of 44–55%. This

Table 9
Carbon balance, normalized to 100 MW of feedstock energy (HHV).

		FB-wood-1	*FB-wood-2*	*FB-wood-3*	*EF-wood*	*HTC-EF-biocoal*
C in biomass	[kg/s]	2.479	2.479	2.479	2.479	2.479
C in natural gas	[kg/s]					0.114
C in liquid HTC byproducts	[kg/s]					0.165
C in gaseous HTC byproducts	[kg/s]					0.088
C to sequestration	[kg/s]	2.093	2.138	2.093	2.125	1.893
C in ash	[kg/s]	0.050	0.050	0.050	0.007	0.007
C released to atmosphere as CO_2	[kg/s]	0.336	0.291	0.336	0.346	0.440
Capture rate[a]	[−]	84.5%	86.3%	84.4%	85.7%	73.0%

[a] Carbon in ash not included.

Table 10
Modelling assumptions and results for the IGCC configurations.

	EF-biocoal-1	*FB-wood-1*	Rhodes [2]
Gasifier type	Entrained flow	Fluidized bed	BCL/FERCO (dual bed, steam blown)
Feedstock	Biocoal, $w = 10\%$	Wood, $w = 50\%$	Not specified
Net electricity output	749.13 MW	137.3 MW	
Gas turbine combustor/expander outlet temperature	1350/605 °C	1231/563 °C	H-class technology
Gas turbine pressure ratio	15.4	14.8	
Live/reheat steam temperatures	580/580 °C	610/610 °C	
Steam pressure levels	127/41/7 bar	110/6 bar	
Condenser pressure	0.05 bar	0.05 bar	
Air separation	Integrated with gas turbine	Not integrated	None
Syngas conditioning	Cold (absorption)	Hot (adsorption)	
Efficiency (HHV), IGCC	33.3%	28.6%	25–28%
Carbon capture rate, IGCC	84.7%	84.5%	44–55%
Efficiency, conversion chain	27.7%		
Capture rate, conversion chain	72.7%		

difference is due to the gasifier type used in [2], where 30% of the feedstock carbon exits the gasifier as char and is later combusted, thereby evading carbon capture. Another 11% of carbon is released to the atmosphere because heat for the steam reformer is provided by the combustion of raw gas rather than low carbon clean gas [2].

5. Discussion

The methane concentration in the raw gas from fluidized bed gasification has been shown to have a strong influence on the efficiency of the syngas production, due to the energy consumption of the downstream steam reformer. For industrial-scale gasifiers, methane concentration will be dependent on the gasifier design and residence time and should be kept as low as possible if CCS is to be applied. Other arrangements for the steam reforming should also be investigated. Burning raw gas rather than clean gas is likely to improve the efficiency but will lead to significant carbon slippage unless the CO_2 can be captured from the combustion gas.

For entrained flow gasification, we used the same gasification temperature and steam-to-feed ratio for bituminous coal, biocoal and wood. Optimizing the gasification temperature and steam-to-feed ratio based on experimental data may improve the cold gas efficiency. Experiments with the gasification of biocoal in an atmospheric entrained flow gasifier have been conducted at Technische Universität München and indicate that the biocoal behaves in a similar manner to lignite (a paper on this work is currently under review).

While we applied hot desulfurization by adsorption for the wood fired fluidized bed gasification, we used absorption at low temperature for the entrained flow gasification of biocoal. If it is certain that only low sulphur biocoal will be used as a feedstock, hot desulfurization could be employed. However, the sulphur content of the biocoal can vary with the biomass feedstock, and a design that accepts both fossil coal and biocoal offers better flexibility. Since the syngas has to be cooled for the CO_2 absorption anyway, the advantage of hot gas desulfurization lies mainly in the possibility to use a clean rather than sour shift.

HTC pre-treatment may provide an interesting option for biomass other than wood which cannot so easily be directly gasified. One consideration for herbaceous biomass is its low ash melting temperature, which can be below 700 °C and may impede fluidized bed gasification [44]. While the gasification of raw biomass requires dedicated systems tailored to the characteristics of the biomass, biocoal can probably be used in generic coal gasifiers, thereby allowing fuel switching from fossil to renewable resources.

In terms of logistics, biocoal, which has a higher energy density than raw biomass, can be produced in several smaller facilities and transported to a central IGCC with carbon capture. This would be especially beneficial for biomass feedstock with a high moisture content to reduce transport costs.

Because of its lower carbon capture rate, HTC is not intrinsically promising for BECCS unless the carbon losses to the liquid phase can be significantly reduced. Since process optimization has not been undertaken, it can be assumed that all the process designs under discussion offer some prospect for improvement. The economic performance of the analyzed systems also needs investigation in order to assess whether HTC plus entrained flow gasification can offer a financial advantage over the direct gasification of raw biomass.

6. Conclusions

Bio-energy with carbon capture and storage (BECCS) is being increasingly discussed because it may lead to negative carbon emissions. This paper investigates three systems of biomass gasification with pre-combustion carbon capture:

- fluidized bed gasification of biomass followed by steam reforming
- direct entrained flow gasification of biomass
- pre-treatment of biomass with HTC and subsequent entrained flow gasification

The gasification of biocoal is more efficient than the gasification of wood, but the losses and auxiliary energy consumption of the HTC process lead to a lower overall efficiency of the conversion from biomass to clean syngas. The efficiency of syngas production with CCS using fluidized bed gasification is strongly dependent on the methane concentration of the raw gas, since this methane needs to be converted in a downstream steam reforming unit to avoid carbon slippage. Up to 9.4% of the feedstock exergy is destroyed in the steam reformer.

The carbon capture rate was found to be 84–86% when wood is directly gasified and 73% when it is first pre-treated with HTC, the lower capture rate being mainly due to carbon losses associated with the gaseous and liquid byproducts of the HTC process. The organic compounds dissolved in the liquid phase of HTC also represent the biggest exergy loss from the HTC plant. HTC research should therefore focus on concepts for limiting these dissolved organic compounds or for converting them into useful products, such as biogas.

HTC followed by a CCS-IGCC with a capacity of 749 MW_{el} and with entrained flow gasification resulted in an overall efficiency from biomass to electricity of 27.7% (on HHV). A CCS-IGCC with direct fluidized bed gasification of the biomass and a capacity of 137 MW_{el} resulted in an efficiency of 28.6%.

All the analyzed biomass to syngas conversion pathways require significant R&D efforts. Direct pressurized entrained flow gasification of biomass is not currently possible because suitable feeding systems are not available. Fluidized bed gasification of biomass has only been proven in units with capacities up to 50 MW_{th} input and experience with pressurized operation and a steam/oxygen mixture as gasification agent is still very limited. Biocoal is likely to work with existing coal gasifiers, but the HTC process itself remains experimental and has only recently reached the point of pilot plant operation.

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